Detection of 15NH2D in dense cores: A new tool for measuring the 14N/15N ratio in the cold ISM

Maryvonne Gerin, Nuria Marcelino, Nicolas Biver, Evelyne Roueff, Laurent H. Coudert, Mohamed Elkeurti, Dariucz C. Lis, Dominique Bockelée-Morvan

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Detection of $^{15}NH_2D$ in dense cores: A new tool for measuring the $^{14}N/^{15}N$ ratio in the cold ISM. ⋆

M. Gerin[1], N. Marcelino[2], N. Biver[3], E. Roueff[4], L. H. Coudert[5], M. Elkeurti[5], D.C. Lis[6], and D. Bockelée-Morvan[3]

[1] LERMA, UMR 8112, CNRS, Observatoire de Paris and Ecole Normale Supérieure, 24 Rue Lhomond, 75231 Paris cedex 05, France. e-mail: maryvonne.gerin@ens.fr
[2] Laboratorio de Astrofísica Molecular, CAB-CSIC/INTA, Ctra de Torrejón a Ajalvir km 4, 28850 Torrejón de Ardoz, Madrid, Spain. e-mail: nuria@damir.iem.csic.es
[3] LESIA, UMR8109, CNRS and Observatoire de Paris, 5, place J. Janssen, 92195 Meudon Cedex. France. e-mail: nicolas.biver@obspm.fr,dominique.bockelee@obspm.fr
[4] LUTh, Observatoire de Paris and UMR8102 CNRS, 5 place J. Janssen, 92195 Meudon Cedex. France. e-mail: evelyne.roueff@obspm.fr
[5] LISA, UMR 7583 CNRS and Université Paris 12, 61 Avenue du Général de Gaulle, 94010 Créteil Cedex, France. e-mail: coudert@lisa.univ-paris12.fr
[6] California Institute of Technology, MC 320-47, Pasadena, CA 91125, USA. e-mail: dcl@caltech.edu

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ABSTRACT

Context. Ammonia is one of the best tracers of cold dense cores. It is also a minor constituent of interstellar ices and, as such, one of the important nitrogen reservoirs in the protosolar nebula, together with the gas phase nitrogen, in the form of N_2 and N. An important diagnostic of the various nitrogen sources and reservoirs of nitrogen in the Solar System is the $^{14}N/^{15}N$ isotopic ratio. While good data exist for the Solar System, corresponding measurements in the interstellar medium are scarce and of low quality.
Aims. Following the successful detection of the singly, doubly, and triply deuterated isotopologues of ammonia, we have searched for $^{15}NH_2D$ in dense cores, as a new tool for investigating the $^{14}N/^{15}N$ ratio in dense molecular gas.
Methods. With the IRAM-30m telescope, we have obtained deep integrations of the ortho $^{15}NH_2D$ $(1_{1,1} - 1_{0,1})$ line at 86.4 GHz, simultaneously with the corresponding ortho NH_2D line at 85.9 GHz.
Results. The ortho $^{15}NH_2D$ $(1_{1,0} - 1_{0,1})$ is detected in Barnard-1b, NGC1333-DCO$^+$, and L1689N, while we obtained upper limits towards LDN1544 and NGC1333-IRAS4A, and a tentative detection towards L134N(S). The para line at 109 GHz remains undetected at the rms noise level achieved. The $^{14}N/^{15}N$ abundance ratio in $^{15}NH_2D$ ranges between 350 and 850, similar to **the protosolar value of ~ 424, and likely higher than the terrestrial ratio of ~ 270.**

Key words. ISM clouds – molecules – individual object (Barnard-1b, L1689N, L134N(S), L1544, NGC1333-IRAS4A) – radio lines: ISM

1. Introduction

Nitrogen chemistry is particularly interesting for understanding the connection between the ISM and the formation of the solar nebula, because it is thought that the primitive atmospheres were nitrogen rich, as Titan remains today. Furthermore, the isotopic $^{15}N/^{14}N$ ratio has been measured in a variety of Solar System bodies, from the giant planets to the rocky planets, comets, and meteorites. The observed differences in nitrogen fractionation are used to understand how these bodies formed within the protosolar nebula. The combination of nitrogen and hydrogen (D/H) isotopic ratios has been demonstrated to be a very effective way of understanding how the ice mantles were enriched in deuterium and nitrogen. Aléon and Robert (2004) have concluded that a fast condensation of the organic matter, enriched in ^{15}N and deuterium, is needed in order to keep a significant fractionation in the solid material of the primitive Solar System. They also have evaluated the exothermicity of the fractionation reactions for nitrogen to be 43 ± 10 K. The D fractionation has not been inherited from the native prestellar core, but most likely occurred in the protosolar nebula

Table 1. Source list

Source	RA (2000)	Dec (2000)	V_{LSR} ($\mathrm{km\,s^{-1}}$)	n(H_2)^a (cm $^{-3}$)
Barnard 1b	03:33:20.9	31:07:34	6.8	3×10^6
NGC1333-IRAS4A	03:29:10.5	31:13:31	7.2	2×10^6
NGC1333-DCO$^+$	03:29:12.3	31:13:25	7.2	1×10^6
L1544	05:04:16.6	25:10:48	7.4	2×10^6
L134N(S)	15:54:08.6	−02:52:10	2.4	2×10^6
L1689N	16:32:29.5	−24:28:53	3.8	2×10^6

[a] From Caselli et al. (2008)

Send offprint requests to: e-mail: maryvonne.gerin@ens.fr
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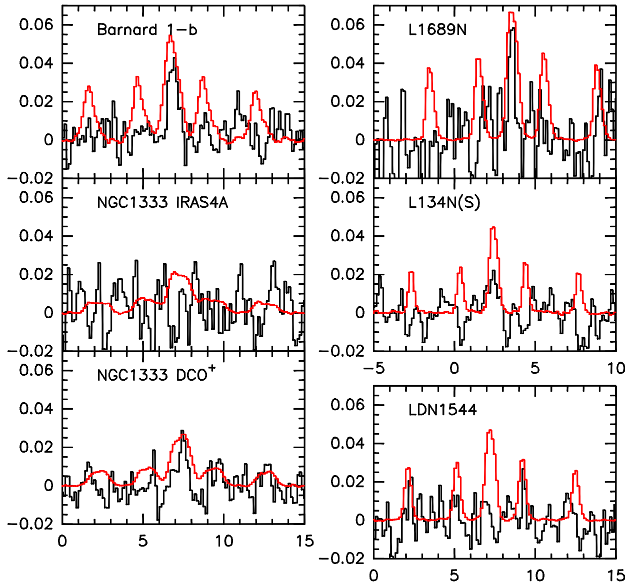

Fig. 1. Spectra of the $1_{1,1}-1_{0,1}$ lines of o-NH_2D (grey/red line) and o-$^{15}NH_2D$(black line) . The vertical scale is $T_{\rm mb}$in K, the horizontal scale is V_{LSR} in $\rm km\,s^{-1}$. The o-NH_2D spectra have been multiplied by 0.02 except for the L1689N spectra that has been scaled by 0.0125.

(author?) (Remusat et al. 2006, Gourier et al. 2008), yet the same physical and chemical processes are thought to operate in the prestellar cores and in the coldest regions of circumstellar disks. In the ISM, recent observations show that, contrary to CO, nitrogen does not deplete from the gas phase in dense cores, except when the density rises significantly above 10^6 $\rm cm^{-3}$. Nitrogen species can therefore be very significantly deuterated, with D/H fractionation of several tenths for N_2D^+ (Daniel et al. 2007; Pagani et al. 2007) and NH_2D (Crapsi et al. 2007). Multiply deuterated ammonia in particular can be very abundant (Gerin et al. 2006; Lis et al. 2002a; Lis et al. 2006; Roueff et al. 2005). Nitrogen molecules will therefore be significant molecular reservoirs of deuterium. It is interesting to study whether they could also be enriched in ^{15}N, and whether signatures from an enrichment at an early evolutionary stage can be identified in primitive matter.

High ^{15}N enhancements are measured both in HCN and CN cometary gases (Bockelée-Morvan et al. 2008; Schulz et al. 2008), **and in primitive carbonaceous meteorites.** High ^{15}N enhancements may have been present in the ammonia ices of the natal presolar cloud according to the fractionation mechanism proposed by Rodgers & Charnley (2008a; 2008b; 2004) and Charnley & Rodgers (2002). Nitrogen fractionation is not expected to be as efficient as deuterium fractionation in dense cores, yet significant departures from the elemental $^{14}N/^{15}N$ ratio may occur in some molecules. As first shown by Terzievia & Herbst (2000), and developed by Charnley & Rodgers (2002) and Rodgers & Charnley (2008a), nitrogen fractionation in the gas phase may operate through ion-molecule reactions involving atomic or ionized nitrogen . Rodgers and Charnley (2008b) have subsequently studied the possible role of neutral-neutral reactions involving ^{15}N and CN. Little observational interstellar data are available. We have therefore started a survey of the main nitrogen-bearing interstellar species in 5 dense cores and a class 0 source (Table 1). This paper reports the detection of o-$^{15}NH_2D$ as the first result of this survey.

Table 2. Einstein coefficients, upper level energies and critical densities for the range of temperatures considered in this work

Molecule	Transition	Frequency (GHz)	A_{ij} ($\rm s^{-1}$)	$E_{\rm up}$ (K)	$n_{\rm crit}$ ($\rm cm^{-3}$)
o-NH_2D	$1_{1,1}-1_{0,1}$	85926.2780	7.82e-6	20.68	4.2 10^6
o-$^{15}NH_2D$	$1_{1,1}-1_{0,1}$	86420.1959	7.96e-6	20.63	4.2 10^6
p-$^{15}NH_2D$	$1_{1,1}-1_{0,1}$	109284.9021	1.61e-5	21.18	8.8 10^6

2. Observations

The microwave and far infrared spectra of $^{15}NH_2D$ and $^{15}NHD_2$ have been recently investigated by Elkeurti et al. (2008) and used to produce the corresponding line lists as supplementary data[1], while accurate line lists and partition functions for the ^{14}N isotopologues of the NH_3 family can be found in Coudert & Roueff (2006). These species are also independently listed in the Cologne Database for Molecular Spectroscopy (CDMS, Müller et al. 2001, 2005), with small differences in the line frequencies due to different handling of the hamiltonians.

We have chosen to search for the $1_{1,1}-1_{0,1}$ line of ortho $^{15}NH_2D$ since the corresponding NH_2D line is very strong and both the sky transmission and telescope performances are excellent **at 86 GHz**. The frequency shift introduced by the ^{15}N substitution is small enough that the two isotopologues can be observed with the same receiver tuning. The line frequencies (Elkeurti et al. 2008), Einstein A coefficients, upper energy levels and critical densities are listed in Table 2. We have used the theoretical estimates of the critical densities from the reduced mass ratio scaling of Machin & Roueff (2006) for the NH_2D–He values at 10 K, the temperature appropriate for the cold cores we have observed. However these values are probably too large when molecular hydrogen is involved, as found in recent calculations of the NH_3-H_2 system by Valiron et al. (private communication).

The observations have been performed with the IRAM-30m telescope, during three observing sessions in December 2007, March 2008 and September 2008. We used the A100 and B100 receivers in parallel, tuned to 86.2 GHz in order to detect o-NH_2D and o-$^{15}NH_2D$ with the same detector setting. The weather conditions were average, with $5-10$ mm of water vapor (PWV) . **The NH_2D and $^{15}NH_2D$ lines were observed simultaneously, with the J=1–0 lines of $H^{15}NC$ and $H^{13}CN$ at 86.055 GHz and 86.338 GHz. We used the VESPA correlator,** tuned to a spectral resolution of 40 kHz, and spectral bandpass of 40 MHz for each line. The data were taken using the wobbling secondary reflector, with a beam separation of 240". Telescope pointing was checked on nearby planets and bright radio quasars and was found accurate to $\sim 3''$. Due to rather poor weather conditions during the September run

[1] Available at `http://library.osu.edu/sites/msa/suppmat/v251.i1-2.pp90-101/mmc1.txt`

(high PWV and cloudy sky), the pointing accuracy was degraded to $\sim 5''$. Additional observations of the p-$^{15}NH_2D$ line at 109.3 GHz were obtained in March 2008. We only searched for this line towards Barnard-1b and detected no signal down to a rms noise level of 18 mK with 0.2 $\mathrm{km\,s^{-1}}$ velocity resolution. For L134N(S), we combined the data with observations performed in April 2005, as part of the dark cloud line survey project (Marcelino et al. 2009). The weather conditions were excellent (1 − 2 mm PWV) and the observations performed in the frequency switching mode.

The data processing was done with the `GILDAS`[2] software (*e.g.* Pety et al. 2005). We used the dec08b version of this software, which allows to correct for a minor bug in the frequency calibration during the observations. The IRAM-30m data are presented in main beam temperatures T_{mb}, using the forward and main beam efficiencies $F_{\rm eff}$ and $B_{\rm eff}$ appropriate for 86 GHz, $F_{\rm eff}$=0.95 and $B_{\rm eff}$=0.78. The **uncertainty in flux calibration** is $\sim$ 10%, as checked by the variation of the intensity of the strong o-NH_2D and $H^{13}CN$ lines in the spectrum. Linear baselines were subtracted.

Because the nuclear spin of ^{15}N is 1/2, the $^{15}NH_2D$ lines are split into fewer hyperfine components than NH_2D which makes their detection more favorable. The hyperfine structure of $^{15}NH_2D$ is driven by the quadrupole moment of the deuterium nucleus, which is much smaller than the corresponding value of ^{14}N. We have checked, by using the nuclear quadrupole constants provided in Garvey et al. **(author?)** (1976), that the resulting hyperfine splitting is less than 50 kHz. We can thus safely assume that the spectrum reduces to a single component . As shown in Figure 1, the $^{15}NH_2D$ line is clearly detected towards Barnard-1b, and L1689N, while we obtained upper limits towards LDN1544 and NGC1333-IRAS4A and **tentative detections towards NGC1333-DCO$^+$ and L134N(S)**. The ratio of peak antenna temperatures **of the NH_2D and $^{15}NH_2D$ lines is 50 - 100, and the velocity agreement is excellent.** Using the JPL and CDMS spectroscopy data bases, we have checked that no line of known interstellar molecules are expected within ±300 kHz from the $^{15}NH_2D$ line frequency. The identification of the detected feature is therefore secure.

The line parameters were estimated by fitting Gaussian profiles to the detected o-$^{15}NH_2D$ lines. For o-NH_2D we used the HFS routine implemented in CLASS, which allows to take into account the hyperfine components self-consistently. The opacity of the ortho NH_2D line is moderate in all sources, with a total opacity for all lines ranging from $\sim$ 1 to $\sim$ 5 (Table 3).

3. Results

3.1. NH_2D and $^{15}NH_2D$

Results of the fits and derived molecular column densities are listed in Table 3. As we are mostly interested in the ratio of column densities, we computed them under the simple assumption of a single excitation temperature. We used the excitation temperature derived from the NH_2D fit for both isotopic species. The o-NH_2D column densities are in good agreement with previously published results for the same sources (Roueff et al. 2005). The [NH_2D]/[$^{15}NH_2D$] abundance ratio range from 360 to 810, with the largest value for L1689N. This last source is an interaction region between a molecular outflow and a dense core, and as such may have peculiar properties (Lis et al. 2002b). Given the error bars, the measured [NH_2D]/[$^{15}NH_2D$] ratio is comparable to the **$^{14}N/^{15}N$ protosolar ratio, as measured in Jupiter (450; Fouchet et al. 2004) and in osbornite-bearing calcium-aluminium-rich inclusions from meteorites (424; Meibom et al. 2007), and likely larger than the terrestrial abundance ratio (270)**. Although the uncertainty on the [NH_2D]/[$^{15}NH_2D$] ratio remains large, the cold prestellar cores L1689N and LDN1544 seem to have a larger ratio than Barnard-1b and NGC1333-DCO$^+$.

3.2. ^{15}N fractionation

Nitrogen fractionation involves two main mechanisms in the gas phase: isotopic dependent photodissociation of molecular N_2, principally at work in the atmosphere of Titan (Liang et al. 2007) and possible ion-molecule fractionation reactions occurring at low temperatures in cold dense cores as first measured by Adams & Smith (1981) and calculated by Terzieva and Herbst (2000). In this latter case, involved endothermicities values range between a few K up to 36 K for exchange reactions involving ^{15}N, $^{15}N^+$, and ^{15}NN. **Selective photodissociation of N_2 and $^{14}N^{15}N$ takes place at wavelengths between 80 and 100 nm, a range where cold dense cores are completely opaque. Then, this mechanism does not work in the present context.** Charley & Rodgers (2002) and Rodgers & Charnley (2008a) have investigated the nitrogen fractionation in their time dependent, coupled gas/solid chemical models. They conclude that ^{15}N-rich ammonia and deuterated ammonia can be frozen onto the ice mantles, provided all nitrogen is not converted into N_2. The gas phase becomes enriched at early times, before the complete freezing of the gas phase molecules.

Additional fractionation reactions may be introduced such as those involving $^{15}N^+$ with CN and NH_3 and some neutral-neutral reactions between ^{15}N and CN (Rodgers & Charnley 2008b). However, none of these reactions has been studied in the laboratory and these schemes remain highly hypothetical. We have developed a gas phase chemical code, including ion-molecule fractionation reactions for carbon and nitrogen (Langer 1992; Langer et al. 1984; Terzieva & Herbst 2000) as well as a complete deuterium chemistry (Roueff et al. 2005). We have explicitly introduced D and ^{13}C on the one hand and D and ^{15}N on the other hand for NH_n, HCN and HNC molecules, in order to directly compare the model results with the observations. The chemical network involves 302 chemical species and 5270 reactions. The maximum number of carbon atoms in a molecule has been limited to 3. We have introduced the additional reaction channels arising from the inclusion of isotopic species. We have also preserved functional groups in dissociative recombination reactions such as :

$$HCND^+ + e \rightarrow HCN + D \quad (1)$$

$$HCND^+ + e \rightarrow DNC + H \quad (2)$$

Note that the branching ratios of the dissociative recombination of N_2H^+ have been measured again by Molek et al. (2007) with the result that the channel towards N_2 occurs with a probability of at least 90%

[2] See `http://www.iram.fr/IRAMFR/GILDAS`

Table 3. Line intensities and molecular column densities

Source	o-NH_2D					o-$^{15}NH_2D$				$\frac{[NH_2D]}{[^{15}NH_2D]}$
	$T_{mb} \pm \sigma^a$	δV	τ	T_{ex}	N^b	$T_{mb} \pm \sigma^a$	I	δV	N^b	
	K	kms^{-1}		K	10^{14} cm^{-2}	mK	$mKkms^{-1}$	kms^{-1}	10^{11} cm^{-2}	
Barnard1b	2.5 ± 0.047	0.79	5.24 ± 0.14	6.0 ± 0.5	4.7 ± 0.5	42 ± 9	30 ± 4	0.67	10 ± 2.7	470^{+170}_{-100}
N1333-IRAS4A	1.0 ± 0.018	1.38	1.39 ± 0.10	5.0 ± 0.5	2.7 ± 0.6	± 10	< 30	...	< 10	> 270
N1333-DCO^+	1.3 ± 0.015	1.15	1.71 ± 0.05	5.3 ± 0.5	2.4 ± 0.4	26 ± 8	14 ± 3	0.52	6.7 ± 2.5	360^{+260}_{-110}
LDN1544	2.3 ± 0.016	0.47	7.05 ± 0.05	5.3 ± 0.5	4.1 ± 0.5	± 7	< 10	...	< 5.2	> 700
L134N(S)	2.2 ± 0.033	0.42	4.75 ± 0.10	5.5 ± 0.5	2.4 ± 0.4	24 ± 7	10 ± 2	0.40	4.5 ± 2	530^{+570}_{-180}
L1689N	5.3 ± 0.030	0.53	6.98 ± 0.02	8.5 ± 0.5	3.4 ± 0.5	65 ± 17	26 ± 6	0.37	4.2 ± 1.5	810^{+600}_{-250}

[a] σ is the rms computed for the original spectral resolution of 40 kHz = 0.136 $km\,s^{-1}$.
[b] computed at LTE with the same T_{ex} for o-NH_2D and o-$^{15}NH_2D$. T_{ex} is derived from the HFS fit of the o-NH_2D profile.

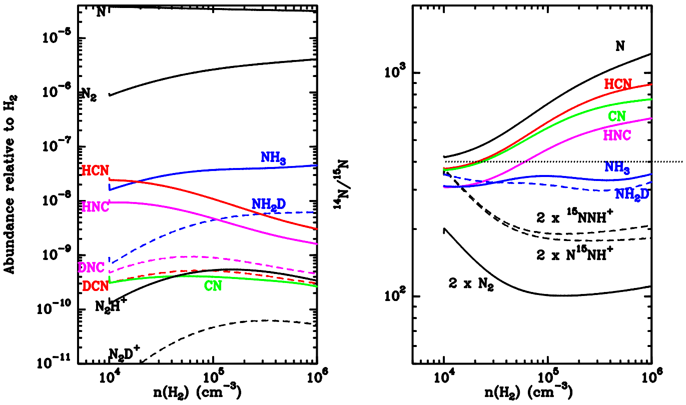

Fig. 2. Prediction of the gas phase abundances relative to H_2 (left) and $^{14}N/^{15}N$ abundance ratio (right) for the main nitrogen species. The models assumes a constant temperature of 10K, and increasing depletions with the gas density, to mimic freezing out. The elemental abundance ratio $^{14}N/^{15}N$ is set to 400.

A calculation is shown in Figure 2 for typical dense core parameters, and assuming a $^{14}N/^{15}N$ abundance ratio of 400, and an ionization rate of $\zeta = 2 \times 10^{-17}$ s^{-1}. The model predicts that the ^{15}N enrichment of ammonia is moderate in the gas phase, while a stronger enrichment is predicted for N_2H^+, and depletion for HCN and CN. Recent models by Rodgers and Charnley (2008a) obtain similar results for the gas phase abundances, the ^{15}N enrichment of ammonia being more efficient in the solid phase.

4. Conclusions

We report the detection of heavy deuterated ammonia, $^{15}NH_2D$, in three cold dense cores. The abundance ratio $[NH_2D]/[^{15}NH_2D]$ is compatible with the $^{14}N/^{15}N$ protosolar value, and seems larger than the terrestrial value despite the remaining measurement uncertainties. While further observations are needed to improve the accuracy and test our chemical models, ammonia and deuterated ammonia seem to be good probes of the $^{14}N/^{15}N$ ratio. Deuterated ammonia is particularly interesting as it probes the coldest and densest regions of prestellar cores which are the reservoirs for the future formation of young stars and their associated protoplanetary disks.

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